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A rapid technique for observation of three-dimensional microstructures: application to analysis of faulted structure in a eutectic alloy

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A RAPID TECHNIQUE FOR OBSERVATION OF THREE-DIMENSIONAL MICROSTRUCTURES: APPLICATION TO ANALYSIS OF FAULTED STRUCTURE IN A EUTECTIC ALLOY

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ABSTRACT

Conventional metallographic techniques, although indispensable for studying the microstructure of alloys, only provide a two-dimensional picture of three-dimensional structures. Hence in the past conclusions regarding the spatial features of microconstituents in opaque materials have been based upon inference from two-dimensional sections, upon three-dimensional models constructed by tedious sectioning procedures, or upon dissolution of one or more phases.

This paper describes a new technique which facilitates the rapid determination of the three-dimensional morphology of phases suspended in an opaque matrix. The rapidity of the technique is based upon the continuous cinephotomicrographic recording of specimen microstructure as the specimen surface is removed under controlled conditions in an electrolytic cell.

The advantages of a rapid technique for determining spatial morphologies in opaque materials are illustrated by application of this method to the study of the configuration of faults in a specimen of $CuAl_2$-Al eutectic alloy. (A fault is the term applied to termination of an extra lamella in the eutectic structure; faults are morphologically similar to

models of edge dislocations in crystals.) A spatial model of the eutectic fault network deduced by the above technique has substantiated the facts that faults in this eutectic alloy tend to run parallel to one another in a direction close to that of the specimen growth axis, and that the majority of faults bridge lamellae rather than lie in a plane parallel to the average lamellar direction. For the specimen area studied, fault density decreased as growth proceeded.

INTRODUCTION

Metallographic observation is a powerful tool for investigating the correlation between structure and properties in metallic alloys. However, ordinary metallographic techniques suffer the fundamental limitation that conclusions regarding the three-dimensional structure of these opaque materials must be inferred from two-dimensional microsections. For this reason it is difficult at best, and frequently impossible to make valid judgments regarding the true spatial distribution of the microconstituents in specimens under investigation.

Various methods have been devised to overcome this basic limitation imposed by conventional two-dimensional microsectioning procedures. Perhaps the technique most obvious is the extension of data obtained on two-dimensional microsections to three dimensions by construction of a spatial model from photomicrographs taken of the same area at different depths within the specimen under consideration. This method has been employed to give some highly interesting and important results[1,2,3]. However, the tedious and time consuming nature of the polish-etch-photograph cycle used to obtain an adequate number of microsections to truly represent a three-dimensional structure in general negates use of this technique when routine analysis of many specimens is desired.

Several authors[4,5,6] have demonstrated that the application of statistical methods to quantitative data obtained from random and/or selected plane sections through opaque material can yield valuable information concerning the volumetric character of such quantities as particle size, shape and distribution, interfacial area and the distribution of linear features. Even though use of these statistical procedures to study the topology of phases would probably be more rapid than construction of a three-dimensional model by the sectioning procedure, it is evident that by their very nature these techniques can give only average values of the quantities studied rather than a detailed physical picture of the microstructure of the phases to be observed.

Rhines and Timpe[7] have suggested that selective dissolution of one or more phases in a multiphase alloy can allow observation of the three-dimensional shape of the undissolved phase(s) and have applied this technique to certain eutectic systems with some success. It is obvious, however, that this method can only be applied to those systems in which one phase is easily removed without subsequent damage to the phase remaining. This limiting condition restricts the wide usage of such a method.

The problem of determining the three-dimensional structure of an opaque material hinges on two facts: the materials under study are opaque to radiation in the visible spectrum and no rapid means of general applicability is available for obtaining the micro-sections necessary to construct three-dimensional models. These facts suggest that the problem may be solved in one of two ways:

(1) The specimen might be bathed with radiation of the correct energy to allow its penetration through the alloy, thus rendering the alloy "transparent", i.e. some variation of microradiography.

(2) A very rapid method for sectioning the material in depth with continuous recording of the microstructure might be devised.

The first approach, microradiography has been utilized with stereographic X-ray photography to discern the internal spatial arrangements of phases in various materials[8,9]. This method, while yielding good results, is limited to quite thin specimens due to the attendant absorbtion of X-rays by metallic alloys. In addition, since data are still recorded upon two-dimensional film some type of projection must be employed to facilitate interpretation of the data. In principle, the first restriction might be circumvented by using more deeply penetrating gamma radiation to replace X-radiation but the relative unavailability of gamma ray sources precludes the use of such a technique. Further, the equipment needed to facilitate the use of X-ray or gamma ray stereomicroradiography is much more complex and expensive than that required for any of the previously described techniques.

Thus the requirements that any technique devised to study the spatial microstructure be <u>rapid</u> and <u>simple</u> with general applicability to observation of the <u>detailed</u> microstructure of <u>various</u> alloys seems to dictate a solution of the problem along the lines of the second approach mentioned in a previous paragraph. Using this approach, a simple new technique which facilitates rapid determination of the three-dimensional morphology of phases suspended in an opaque matrix was developed. Continuous cinephotomicrographic recording of specimen microstructure as the specimen surface is removed at a controlled rate in an electrolytic cell provides such a rapid and simple technique based upon fundamental metallographic principles. The practicality of similar methods has been demonstrated recently by studies of the three-dimensional nature of dislocation loops in Zn[10] and spheroidized structure in steel[11].

DESCRIPTION OF APPARATUS

The apparatus designed for the three-dimensional studies is shown in Figure 1 and depicted schematically in Figure 2. It consists of an American Optical Model 2200 triocular microscope body and lens system which have been adapted to allow continuous cinephotomicrography of a specimen microstructure while its surface is removed by electrolytic dissolution in a plexiglass cell fixed to the microscope stage. The specimen to be studied is inserted in a special plexiglass mount (see details of electrolytic cell, Figure 3) which contains an aluminum electrical contact and a nozzle (containing a slit 1/64" high by 1/2" wide) adjusted to allow the electrolyte to flow evenly over the specimen surface during electropolishing. The mount is then screwed to the base of the cell which contains the electrolyte during the polishing operation. A two-inch square of 1100 aluminum with a one-inch diameter hole drilled through it to allow passage of the objective lens forms the cathode of the electrolytic cell, the specimen being the anode. The cell is traversed vertically to focus the objective lens upon the specimen surface within the solution by means of a calibrated knob controlling the microscope stage. The specimen can be continuously observed through the binocular eyepieces of the microscope and its microstructure simultaneously recorded by a Keystone Model A 16 mm movie camera mounted on the vertical tube of the triocular head. During electrolytic removal of the specimen surface, the electrolyte is continuously circulated through the plexiglass cell to remove "corrosion" products from the specimen surface as they are formed and to avoid overheating of the electrolyte. Recirculation is carried out by allowing excess fluid to overflow the cell, pumping it up to a reservoir of fixed head and allowing the fluid to return by gravity feed to the cell. The input flow rate can be controlled by a valve in the feed line.

In practice the specimen to be surveyed, after being belt ground and polished on 1/o paper, was fixed to the electrical contact on the plexiglass mount and all surfaces with the exception of that to be studied were coated with Micromask, an inert lacquer. The electrolyte consisting of one part glycerol, one part perchloric acid and seven parts ethanol was introduced into the cell and the circulation system activated. A 20X objective lens was focused upon the specimen surface (this lens was chosen as the best compromise giving both sufficient working distance to allow smooth flow of electrolyte over the specimen and the degree of resolution necessary to facilitate detailed study of the eutectic microstructure) and the voltage which had been previously set to give 0.2 to 0.3 amps/cm^2 current density was applied across the cell terminals by means of a potentiometer. The cathode was maintained at a distance of about one inch from the specimen at all times during the electropolishing.

It was observed that upon initial application of the voltage the specimen became covered with a dense black film but within one minute the action of the fluid flow across the specimen surface removed this film revealing a clearly defined microstructure free from any scratches incurred during the initial grinding and polishing operations.

While the specimen surface was being removed at a controlled rate, the distance traversed into the specimen could be noted from divisions on the fine adjustment of the microscope focus knob. Since this was calibrated, the exact distance between succeeding layers of the microstructure could always be determined. Photomicrographs were taken at any desired depth simply by activating a switch on the 16 mm camera which caused short lengths of film to be exposed at a framing rate of 10-20/sec. In this way photomicrographs at 5 μ

intervals over a total distance of 800 μ specimen depth were obtained in approximately an hour, an achievement which would probably have taken many man-hours by normal polishing and etching methods. From these photomicrographs the three-dimensional distribution of microconstituents could readily be obtained.

EXAMPLE OF APPLICATION OF TECHNIQUE

The Al-$CuAl_2$ eutectic was chosen for study since its relatively low eutectic melting point facilitates experimental work, and in the hopes that information derived from a careful study of the spatial configuration of its phases would yield information that could be correlated with the extensive data[3,6,12,13,14] that has already been compiled concerning the solidification and crystallography of this eutectic.

The master heat for these experiments was made by induction melting 99.999$^+$% Cu and 99.994% Al and casting into a magnesia crucible under vacuum. From this ingot specimens were cut by band saw for subsequent uni-directional solidification. These eutectic specimens were directionally solidified horizontally in a graphite crucible at a rate of 0.5 cm/hr. This rather slow growth rate was chosen for these preliminary experiments in order to produce relatively thick lamellae (according to the well known relation between lamellar spacing and growth rate, $\lambda^2 R$=constant)[13] which would be easily discernible under the microscope. The microstructure of these directionally grown ingots consisted throughout of lamellae that were substantially parallel to one another.

A specimen was removed from one of these ingots at a distance of 1 1/2 inches from the head end, belt ground and fixed in the apparatus as described above. The specimen was cut so that a transverse section of the ingot was exposed to the electrolyte and polishing was then initiated in a direction parallel to the ingot growth direction. After focusing the objective lens on an area within a single eutectic "grain" of the sample, the camera was activated for one second (exposing about 20 16 mm frames) at half minute intervals while the specimen surface was continuously removed. Electrolysis was continued for a period slightly over an hour during which time over 770 μ of specimen were traversed, photomicrographs being taken at approximately 5 μ intervals. A selected frame from the twenty taken at each exposure was printed directly from the 16 mm film using an Enlahead lens-condenser system in conjunction with a Simon Omega enlarger to give negative prints at a magnification of about 650 X.

The faulted structure first described by Kraft and Albright[3] in $CuAl_2$-Al eutectic and later observed in the microstructure of other eutectic alloys[15,16] undoubtedly plays a significant role in the solidification of these kinds of alloys. For this reason it was decided that some insight into the eutectic solidification process might be gained by undertaking a careful investigation of the three-dimensional distribution of faults in a eutectic alloy. In order to facilitate this analysis a three-dimensional model of the faulted structure in the eutectic grain studied was constructed by use of photomicrographs obtained during the continuous polishing process.

From the total sequence of photomicrographs taken, particular photomicrographs corresponding to sections at 24 μ intervals in the specimen were chosen for detailed study (see representative sequence in Figure 4). Employing the procedure outlined by Kraft and Albright[3] faults on each photograph were marked using two different colors to differentiate positive from negative faults. The locations of the faults on each photograph were then transferred with the aid of a properly scaled coordinate system to 1/16" x 12" x 12" plexiglass sheets. Holes were drilled through the marks locating each fault and the sheets were then inserted into a frame so that each sheet was separated by a distance proportional to the distance between those microsections in the actual specimen which they represented. When a wire was passed through the holes on succeeding sheets corresponding to a particular fault, the path of the fault through the volume of eutectic specimen was represented. Wires of different colors were used to distinguish positive from negative faults. By representing all the faults passing through the area of eutectic grain studied in this manner, the three-dimensional model was completed. Figures 5 through 10 are photographs of the three-dimensional model so constructed. It should be noted that in order to restrict the model to a convenient size the vertical scale was compressed so that the ratio of distance measured on the vertical scale to that on the horizontal scale is 0.75 to 1.0.

DISCUSSION OF RESULTS

Careful scrutiny of the three-dimensional model of the fault network in the specimen investigated combined with detailed studies of the photomicrographs from which the model was made lead to the following conclusions regarding the fault structure in the specimen.

First, the faults did not assume a random array in space but tended to lie apporximately parallel to the same direction. This gross fault direction lay close to the specimen growth direction as can be seen in pictures of the front and side view of the model, Figures 5 and 6. (Some faults near the top and bottom of the model in these photographs appear branched due to their reflection in the plexiglass sheets.) The tendency for faults to lie nearly parallel to the growth direction becomes even more apparent when it is recalled that the vertical scale of the model has been compressed somewhat. These observations confirm those previously obtained by Kraft _et al._[6] who applied a statistical analysis to data obtained on a $CuAl_2$-Al eutectic specimen grown under similar conditions.

Secondly, it was observed that faults showed a greater tendency to cross lamellae than run parallel to them as growth proceeded. This is especially evinced by the presence of several isolated fault loops in the model (An example of such a loop is indicated by the arrow in Figure 5 and shown close up in Figure 7.). In general, these loops were nearly planar and the planes (of the loops) were observed to be perpendicular, or nearly so, to the actual lamellae. This implies that faults in these loops lie along "mismatch surfaces", which cross lamellae, rather than lying in planes parallel to the lamellae.

A further illustration of this tendency for faults to bridge lamellae rather than run parallel to them as growth proceeds can be seen by referring to Figures 8, 9 and 10. Figure 8 is a top view of the model; the average lamellar direction in this transverse section runs from lower left to upper right (arrow). The white "line" shown on the photograph lying perpendicular to this direction is the photographic image of a tape placed on the interior of the model. It will be noted that no faults "cross" this tape. Furthermore, it would be impossible to place the tape parallel to the lamellae (arrow) and observe the same phenomenon. Photographs of longitudinal sections of the model taken perpendicular and parallel to the mean lamellar direction are shown in Figures 9 and 10. In Figure 9 a similar white tape on the side of the model opposite to that photographed defines a border across which faults do not* meander as growth proceeds. No such boundary could be established in Figure 10. Taken collectively, these three photographs reveal a pronounced tendency for faults to cross lamellae rather than run parallel to them as growth proceeds. The foregoing conclusions have been recently confirmed by an entirely different type statistical study conducted by Thomas and Kraft[17] on other specimens of the $CuAl_2$-Al eutectic.

A third fact discerned from consideration of the model was that as growth proceeded along the ingot it appeared to become more perfect in structure. That is, there was a decrease in fault density as growth took place.

It has been shown from annealing experiments[18] that spheroidization of lamellar $CuAl_2$-Al eutectic specimens is initiated at faulted regions in the structure and hence it is logical to assume that these are areas of

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*At the bottom of the photograph in both Figures 8 and 9 one "black" fault "crosses" the tape. However, it will be noted that if the tape were bent slightly to the right, the tape would then separate the model into two portions and no faults would cross the boundary.

higher free energy than the surrounding regions of perfect lamellarity. The fact that fault density appeared to decrease as growth proceeded may be evidence that the eutectic structure could obtain a lower energy configuration by the elimination of these higher energy faulted regions through complex fault interactions. Such cancellation of positive and negative faults was observed within the spatial model. However, due to the fact that only several hundred microns of specimen were traversed in this study and the area under observation was small, it would be presumptuous to draw any hasty conclusions regarding this trend without first performing more quantitative statistical studies of fault density along the length of a unidirectionally solidified eutectic specimen.

An interesting example of the results of fault interactions can be seen in Figure 4. Two "mismatch surfaces" (denoted by arrows in picture A) are seen to grow together (B to F) as growth proceeds along the ingot until they cancel one another to produce a region of more perfect lamellarity containing one net fault (G). A similar process takes place for a second pair of mismatch surfaces denoted in picture B which grow together to form a loop in H. Such behavior is further evidence of the tendency for the structure to become more simplified as growth proceeds.

The results obtained here based directly upon the observation of a three-dimensional model of the fault network in a eutectic specimen and recently confirmed by the statistical studies of Thomas and Kraft[17] seem--at least partly--to be in contradiction with one of the fundamental hypotheses of the theory of eutectic growth proposed by Jackson and Chalmers[19]. They have assumed that the stability of faults is the criterion for stable growth of a lamellar structure. They propose that a change in lamellar spacing is accounted for by movement of faults in planes parallel to the lamellae resulting in the "squeezing-in" of extra lamellae, if growth rate increases, or the "squeezing out" of lamellae, if the rate decreases. The results obtained here, however, show that, in the main, faults tend to bridge lamellae, thus suggesting that a fault network may not be an essential feature of lamellar growth. Perhaps, then, as Thomas and Kraft[17] suggest, a "fault-free" structure can be grown by solidifying an ingot in such a way as to prevent new fault loops from initiating and permitting existing ones to grow out.

SUMMARY

1. A rapid method for surveying the three-dimensional shape and morphology of phases suspended in an opaque matrix has been developed.

2. Application of this technique to a unidirectionally solidified $CuAl_2$-Al eutectic has revealed the following facts concerning lamellar fault structures:

a) faults are essentially parallel to one another and the average fault direction is approximately parallel to the specimen growth axis;

b) for the most part faults tend to cross lamellae rather than lie in a plane parallel to the lamellae;

c) fault density decreases as growth proceeds for the specimen studied.

3. This technique for determining the three-dimensional structure of alloys should make possible a systematic and detailed study of the role of the faulted structure in eutectic alloy solidification. In particular this technique should yield interesting results when applied to studies of fault structure in eutectics grown with specific orientation and under controlled conditions probably necessary to produce a structure of reduced fault density.

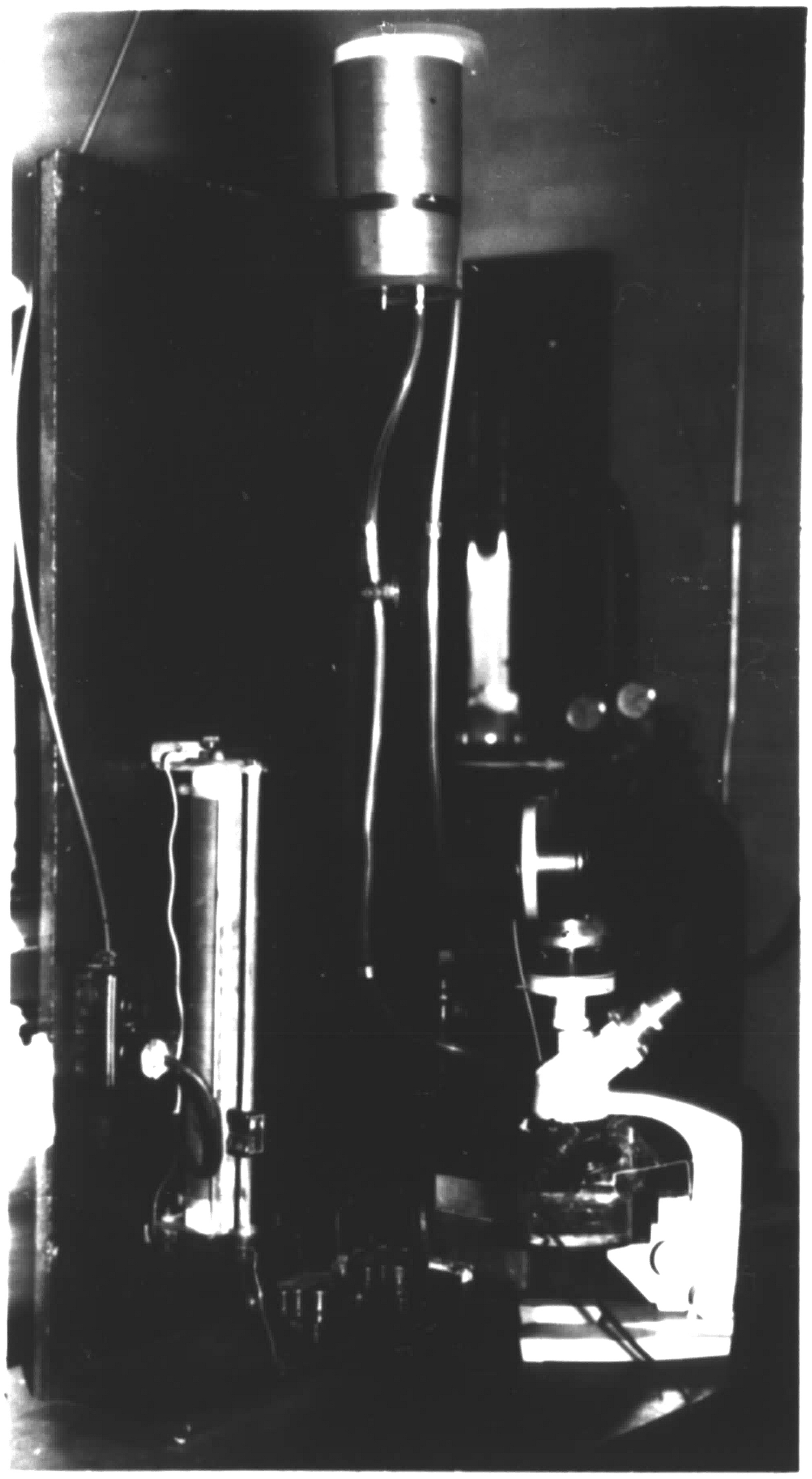

Figure 1. Photograph of apparatus.

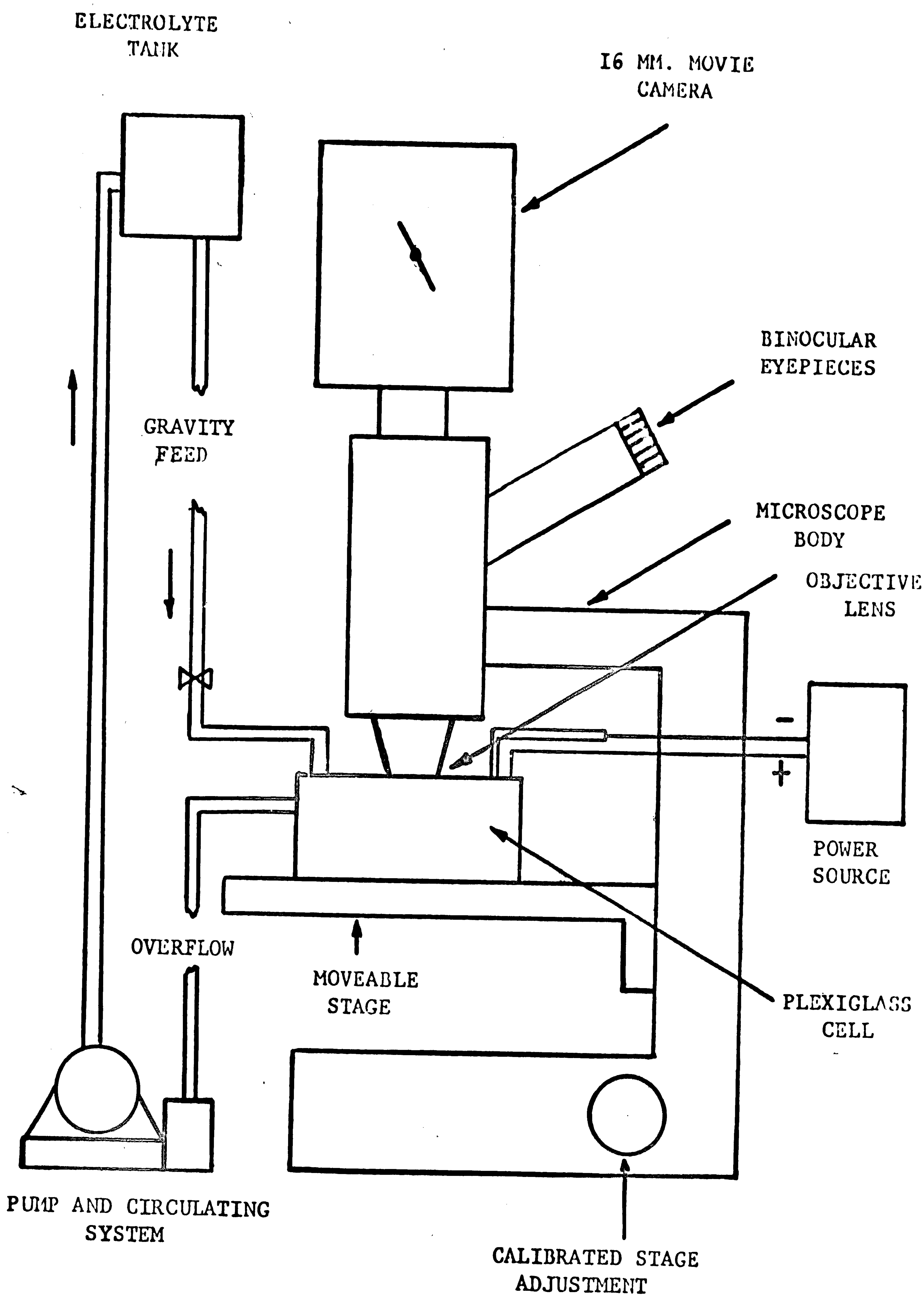

Figure 2. Schematic diagram of apparatus depicted in Figure 1.

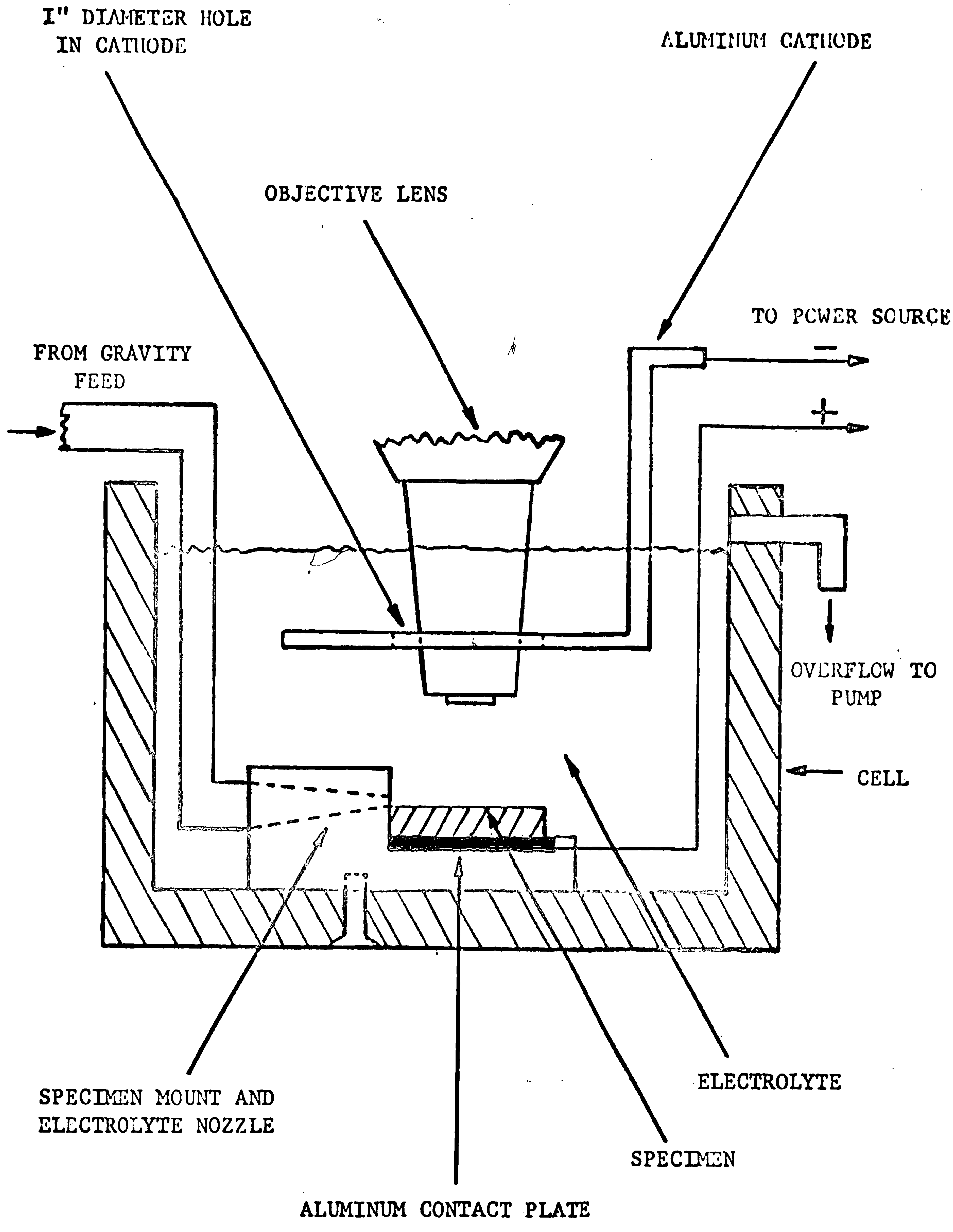

Figure 3. Schematic diagram illustrating cross section of electrolytic cell design.

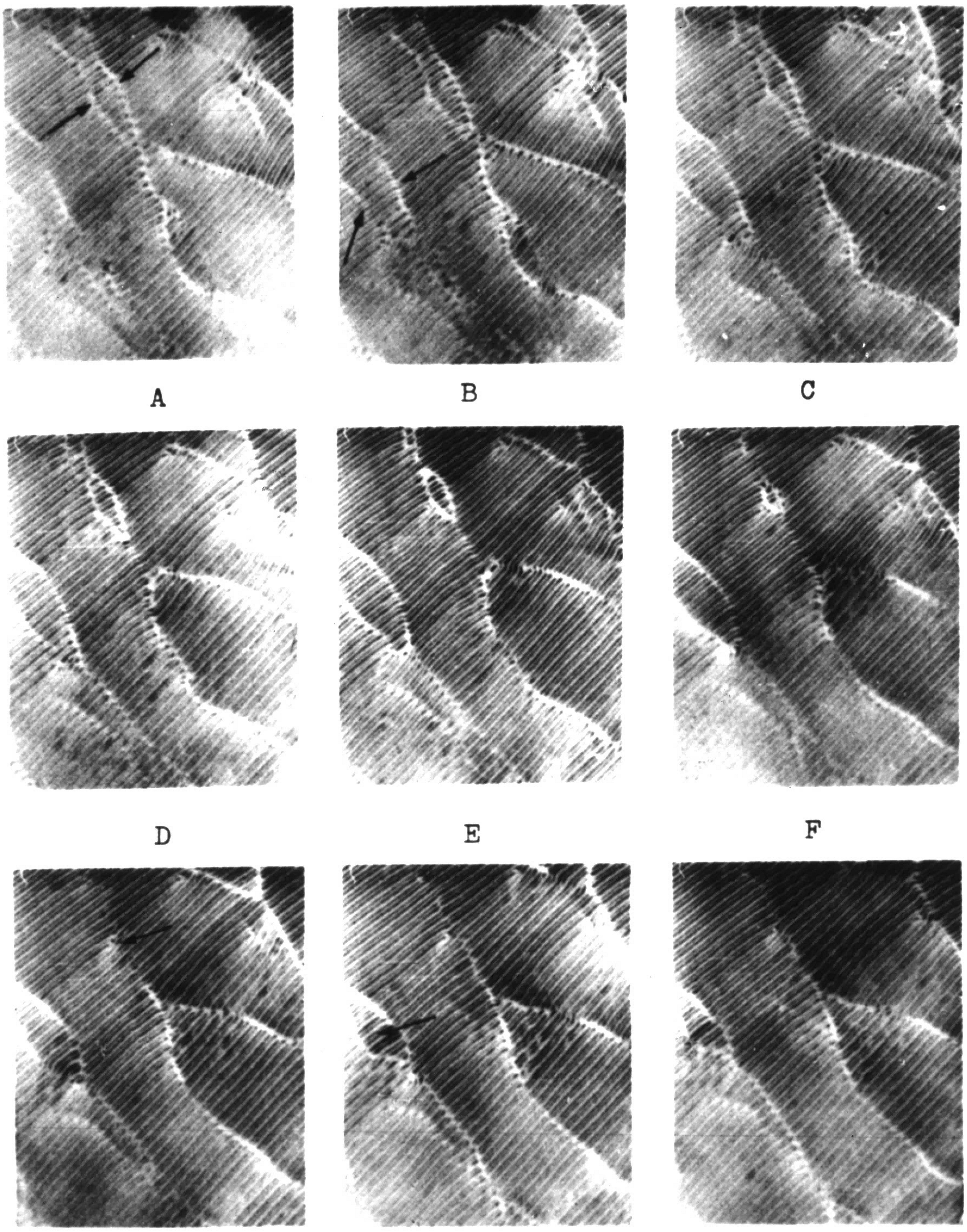

Figure 4. Sequence of photomicrographs representing successive sections in the specimen at about 10 μ intervals. Growth direction from A to I. X 650. Reduced approximately 50 per cent for reproduction.

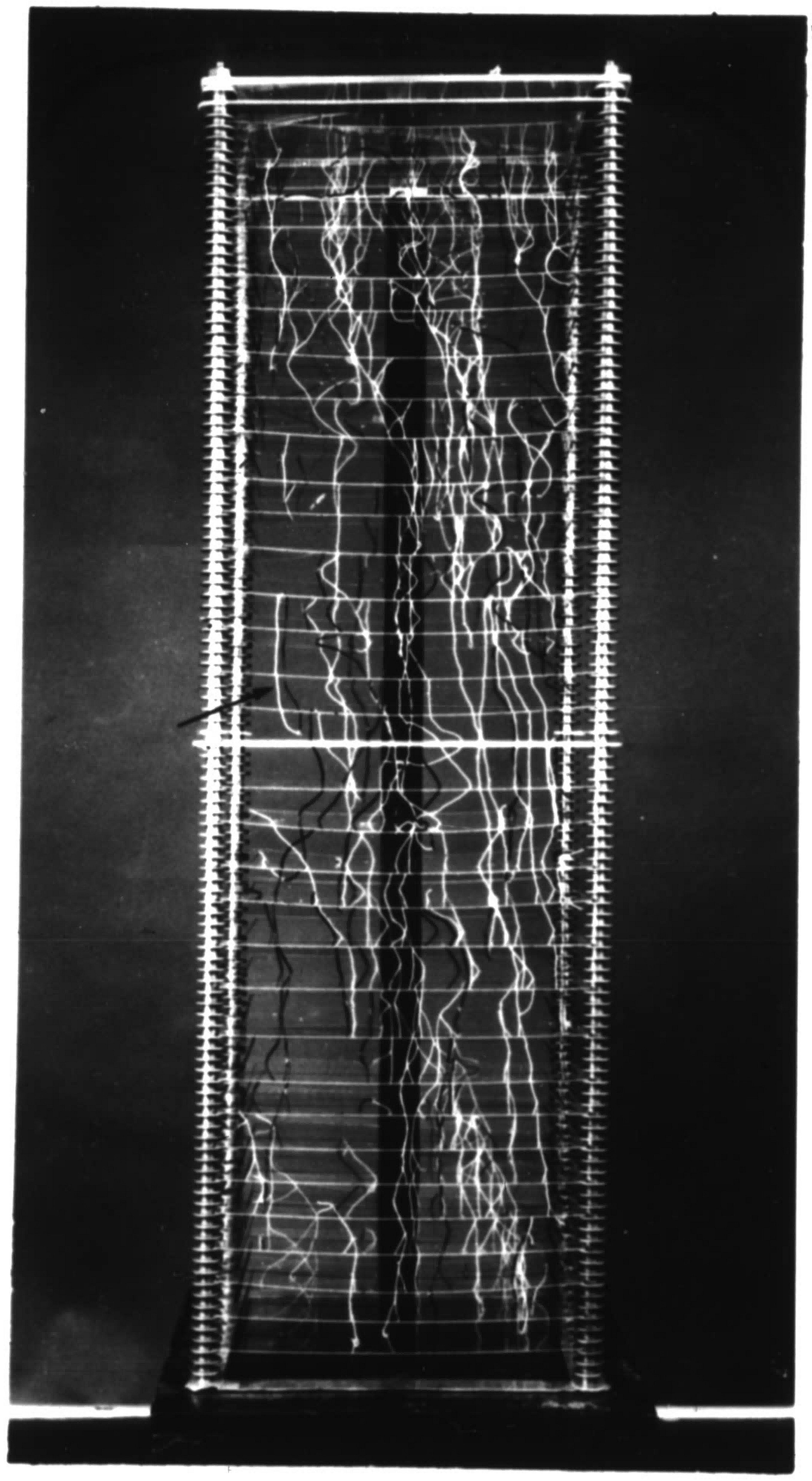

Figure 5. Photograph of front view of the three-dimensional model of fault networks in a $CuAl_2$-Al eutectic alloy. Arrow indicates an isolated fault loop.

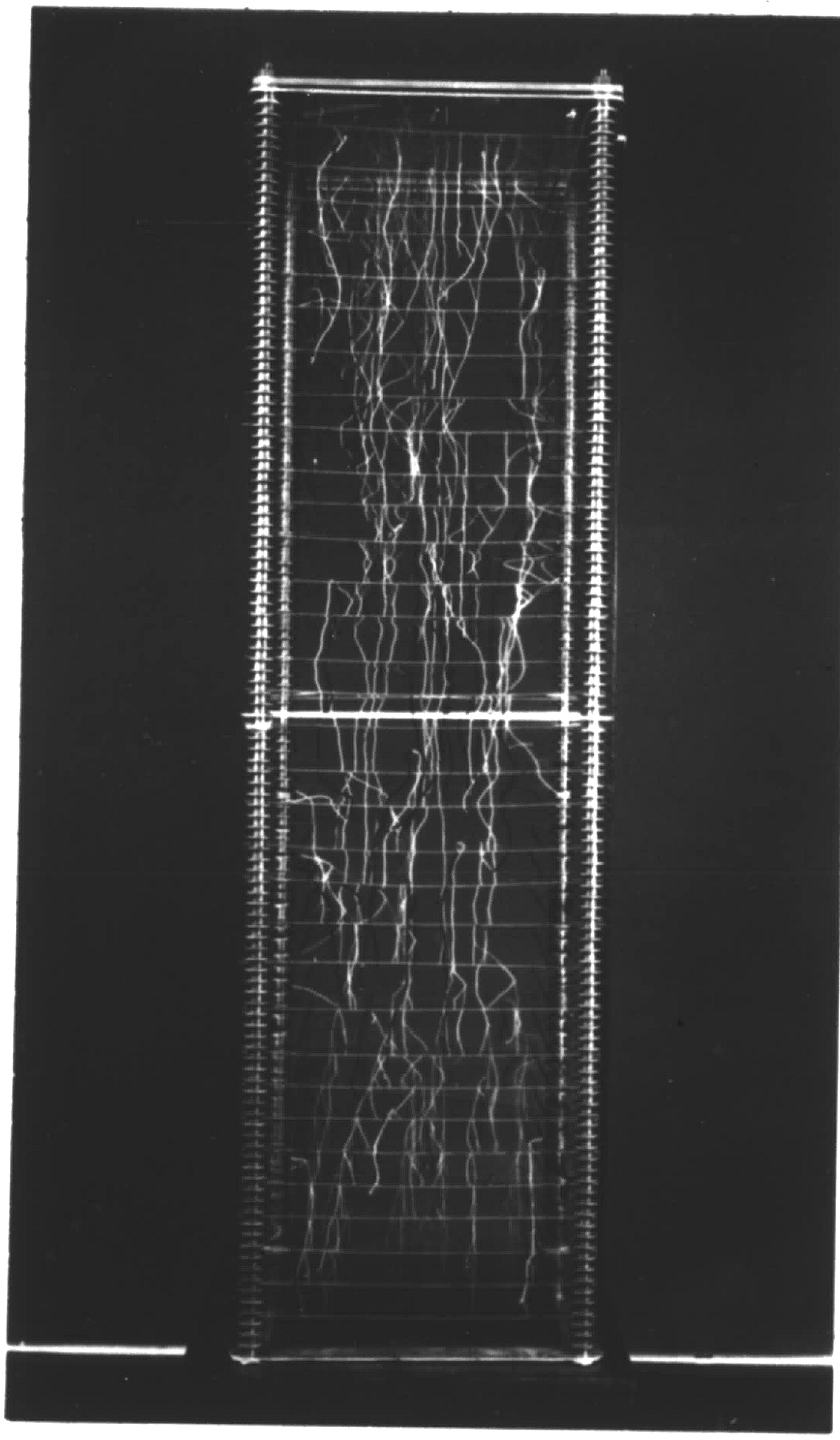

Figure 6. Photograph of side view of model.

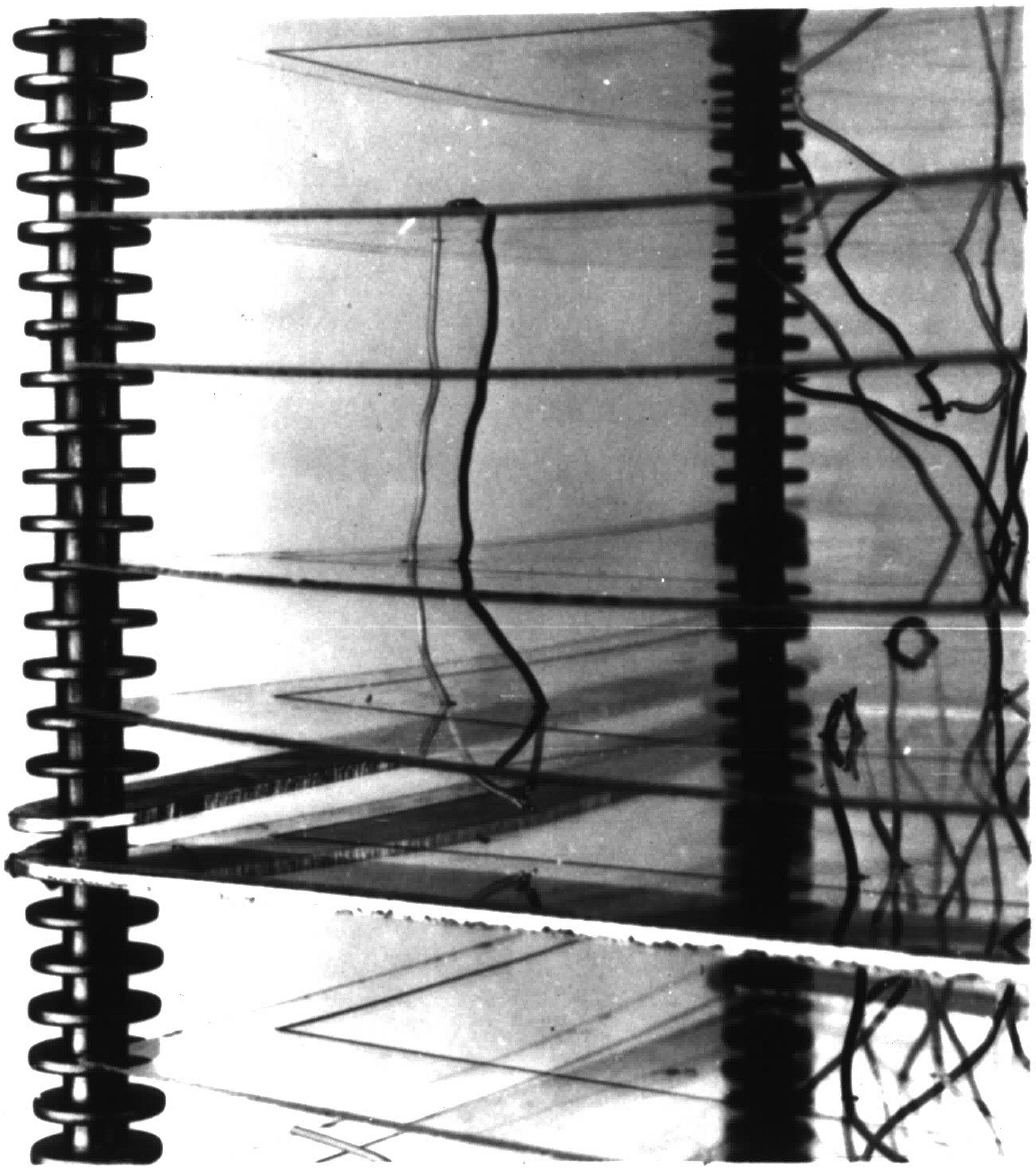

Figure 7. Detail of fault loop indicated by arrow in Figure 5.

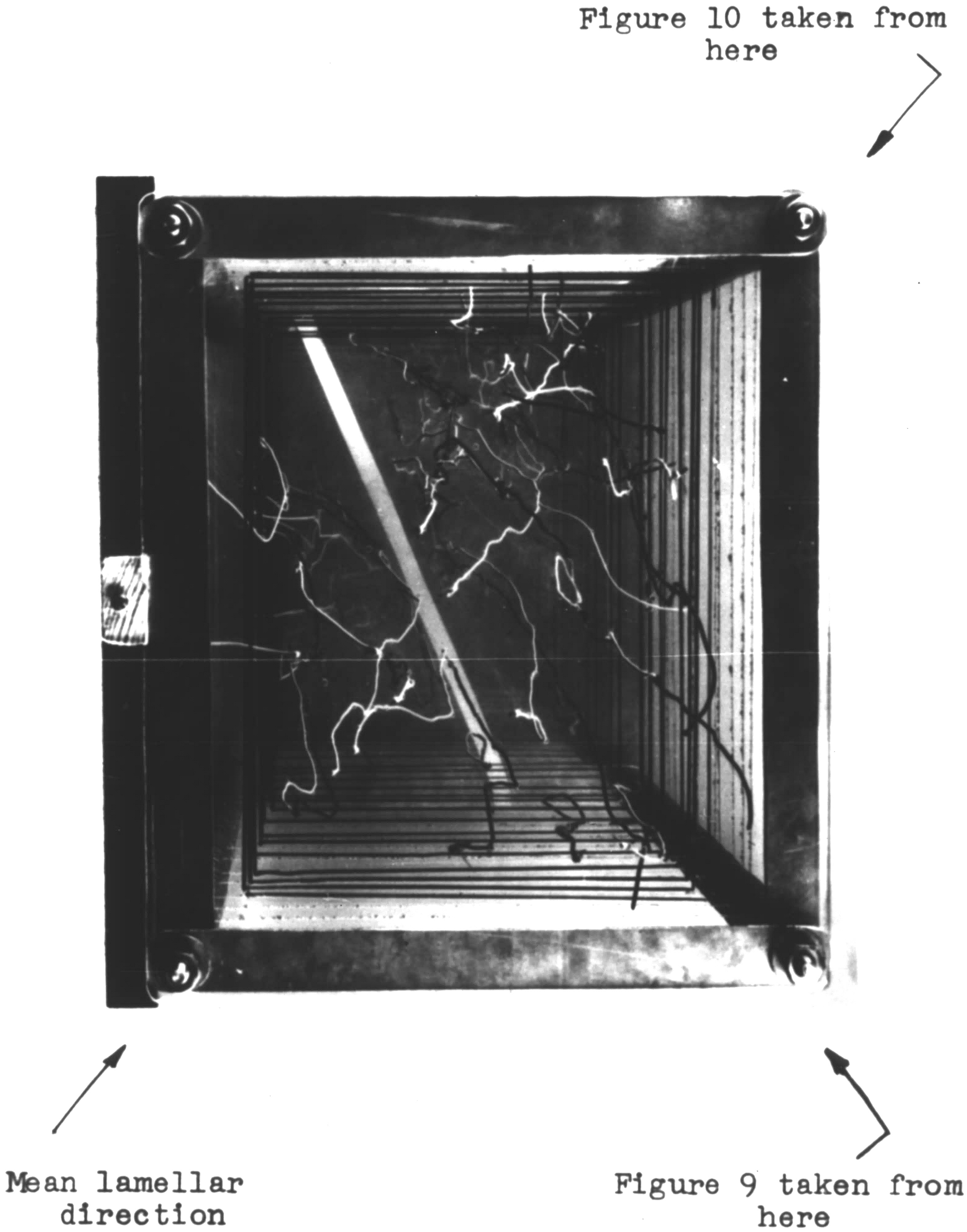

Figure 8. Photograph of top of model. Transverse section. Arrows indicate mean lamellar direction and positions of photographs shown in Figures 9 and 10.

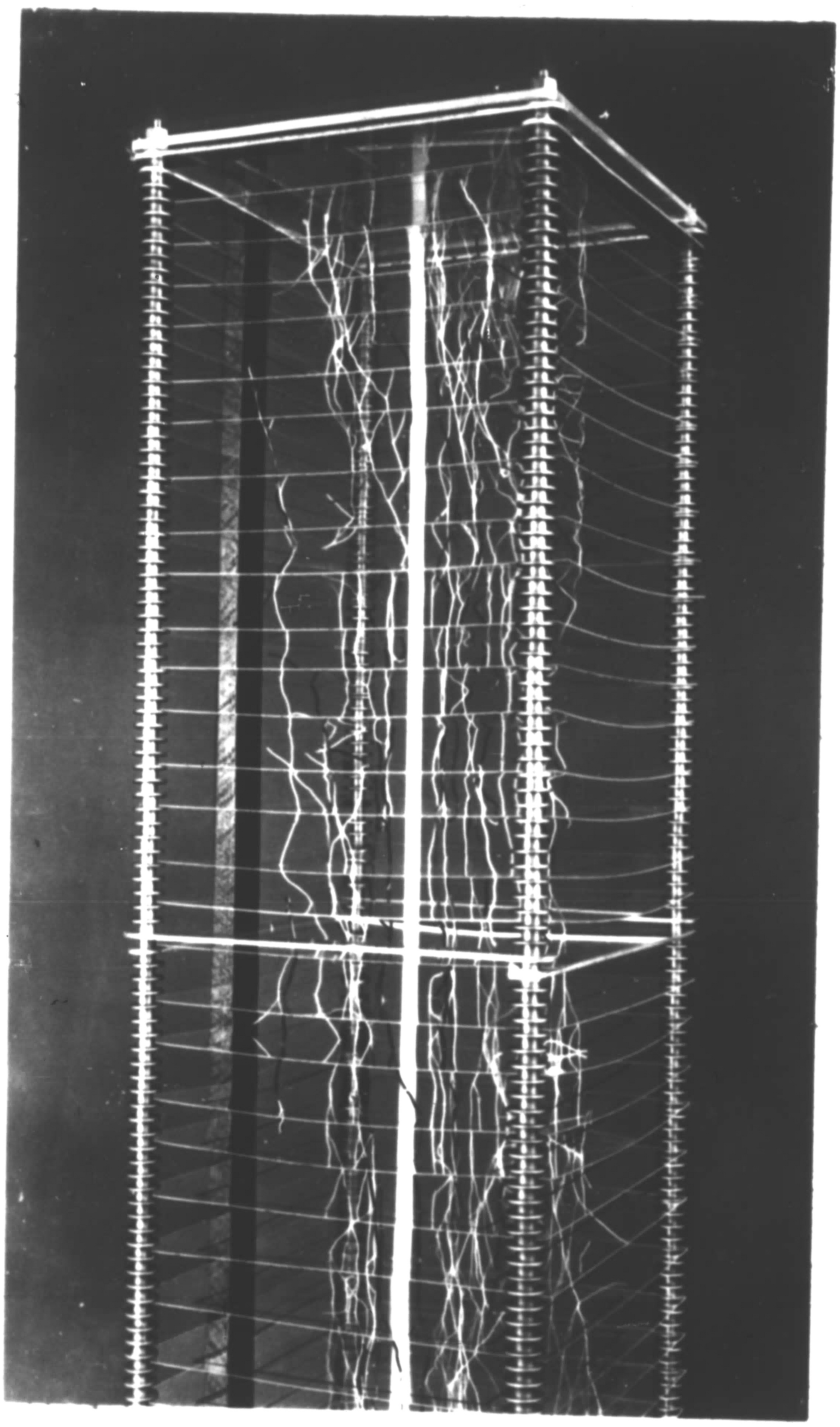

Figure 9. Photograph of longitudinal section of model perpendicular to the mean lamellar direction.

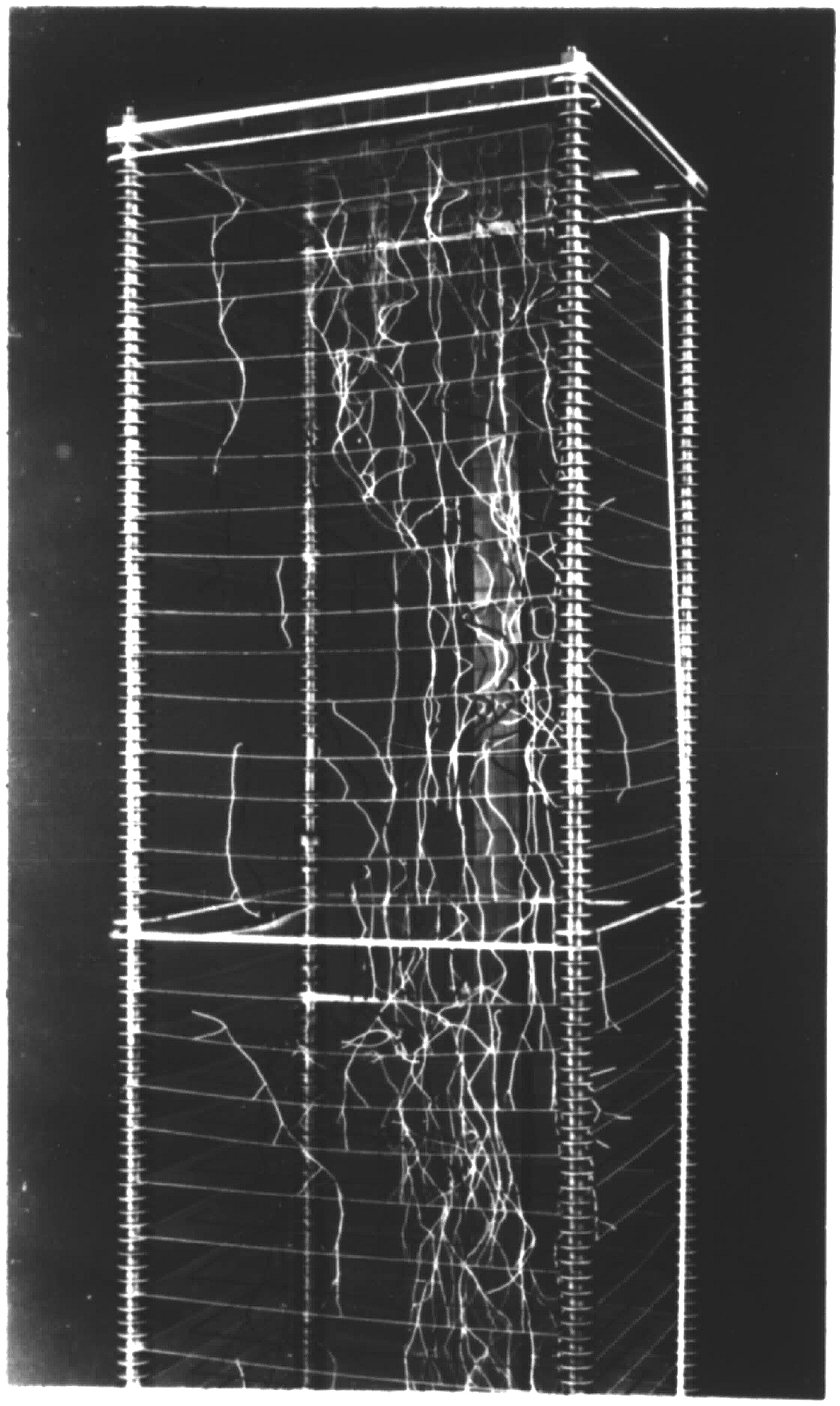

Figure 10. Photograph of longitudinal section of model parallel to mean lamellar direction.

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VITA

Richard Hopkins, son of Mr. and Mrs. Reginald F. Hopkins, was born in Hartford, Connecticut, December 23, 1940. He received his elementary and secondary education in the school system of Woodbury, New Jersey. Upon graduation from Woodbury High School he entered Lehigh University in the fall of 1959 on a scholarship.

As an undergraduate, Mr. Hopkins was a member of Theta Xi social fraternity as well as Phi Eta Sigma and Tau Beta Pi national honor societies. In 1963 he received his B.S. in Metallurgical Engineering graduating with honors.

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